

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Application No.: 10/763,633
Filing Date: January 22, 2004
Applicant: Susan G. Yan
Group Art Unit: 1745
Examiner: Keith D. Walker
Title: MEMBRANE ELECTRODE ASSEMBLY PREPARED
BY DIRECT SPRAY OF CATALYST TO MEMBRANE
Attorney Docket: GP-303571

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APPELLANT'S APPEAL BRIEF

This is Appellant's Appeal Brief filed in accordance with 37 CFR § 41.37 appealing the Examiner's Final Office Action mailed July 16, 2007. Appellant's Notice of Appeal was filed on October 11, 2007. The Appeal Brief fee is enclosed.

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I. Real Party in Interest

The real party in interest for this appeal is the General Motors Corporation of Detroit, Michigan, the assignee of this application.

II. Related Appeals and Interferences

There are no related appeals or interferences.

III. Status of the Claims

Claims 1, 2, 5-8, 10-14 and 21-29 are pending in this application, and are on appeal. Claims 1, 2, 5-8, 10-14 and 21-29 stand rejected. Claims 3, 4, 9 and 15-20 have been cancelled. No claims have been allowed. No claims stand objected to.

IV. Status of Amendments

All amendments have been entered.

V. Summary of Claimed Subject Matter

Independent claims 1, 13 and 21 are all directed to a method for fabricating a membrane electrode assembly (MEA), such as fabricating the MEA 24 shown in figure 1 and described in paragraphs [0023]-[0026] of the specification. Figure 2 is a flow chart diagram showing various steps in the process of fabricating the MEA 24, as described in paragraphs [0028] and [0029], page 7, lines 13-30. All of the independent claims 1, 13 and 21 include the step of providing a proton conducting membrane, such as membrane 14, page 6, lines 6-8. All of the independent claims 1, 13 and 21 further include the step of spraying a catalyst ink on the membrane, such as by using a sprayer 16 to spray a catalyst ink 18 on the membrane 14 to form a catalyst layer 12 on one side of the membrane 14 and a catalyst layer 28 on the other side of the membrane 14, see

paragraphs [0023] and [0025], page 6, lines 8-12 and lines 23-25. Independent claim 1 further includes the step of clamping the membrane 14 in a clamp 32 (originally clamp 28, but changed to clamp 32 during prosecution) to prevent the membrane 14 from wrinkling as a result of the wetness of the catalyst ink 18 on the membrane 14, see paragraph [0024], page 6, lines 16-18. Independent claim 13 further includes the step of spraying the catalyst ink 18 over several passes to deposit the catalyst ink 18 on the membrane 14 to the desired thickness, see paragraph [0026], page 6, lines 27-32. Independent claim 13 further includes the step of drying the MEA 14 under a heat lamp 30 as the catalyst ink 18 is sprayed during the several passes to dry the catalyst layer 12 or 28, paragraph [0024], page 6, lines 18-20 and paragraph [0026] page 6, line 27-29. Independent claim 21 further includes that the catalyst ink 18 has a catalyst, solvent and a ionomer having a concentration that is about half of the ionomer concentration of the catalyst as a ration of ionomer to carbon in a final cathode or anode of the MEA 24, see paragraph [0027], page 7, lines 8-12.

VI. Grounds of Rejection to be Reviewed on Appeal

Whether claims 1, 2, 5-8, 10, 11, 13, 14 and 21-29 should be rejected under 35 USC 103(a) as being unpatentable over U.S. Patent Application Publication 2002/0034675 to Starz (hereinafter Starz);

Whether claims 2, 6, 12-14, 22-24 and 29 should be rejected under 35 USC 103(a) as being unpatentable over Starz in view of U.S. Patent No. 6,277,513 issued to Swathirajan (hereinafter Swathirajan); and

Whether claims 5, 12, 13, 23 and 29 should be rejected under 35 USC 103(a) as being unpatentable over Startz in view of U.S. Patent No. 5,330,860 issued to Grot (hereinafter Grot).

VII. Argument**A. Claims 1, 2, 5-8, 10, 11, 13, 14 and 21-29 are not obvious in view of Starz****1. Independent claims 1, 13 and 21**

Independent claim 1 is a method for fabricating a membrane electrode assembly (MEA) and includes spraying a catalyst ink on a proton conducting membrane to deposit a catalyst layer, where the membrane is clamped to prevent the membrane from wrinkling as a result of the wetness of the catalyst ink on the membrane.

Independent claim 13 is also a method for fabricating an MEA, and includes spraying a catalyst ink on the membrane to deposit a catalyst layer over several spraying passes, where the MEA is being dried under a heat lamp as the catalyst ink is being sprayed on the membrane over the several passes.

Independent claim 21 is also a method for fabricating an MEA and includes spraying a catalyst ink on a proton conducting membrane where the catalyst ink includes a catalyst, solvent and an ionomer having a concentration that is about half of the ionomer concentration of the catalyst as a ratio of ionomer to carbon in a final cathode or anode of the MEA.

2. Starz

Starz discloses a process for producing membrane electrode assembly's including noble metal-containing nanoparticles. Paragraph [0031] states that, "the preparation with the nanoparticles, optionally mixed with other additives such as, for example, dissolved ionomer, carbon black or further electrocatalysts, is applied to the membrane in a spray process, by brushing or immersing or by means of screen

printing.” Paragraph [0054] states, “the resulting ink was applied in a spray process to the front and rear faces of a Nafion membrane” Paragraph [0054] also states, “[t]hen drying was performed at temperatures of 80°C in a circulating air oven.”

3. Discussion

The Examiner states on page 3 of the Office Action that Starz is silent to clamping the membrane, but it would be obvious to use a clamp to hold the membrane in the process of making an MEA. The Examiner further states that Appellant admits this on page 8 of the arguments stating that, “one of ordinary skill in the art would readily recognize what type of clamp would be needed for this purpose. . . .” Appellant respectfully states that this statement in the Arguments section of the Response to Office Action is not an admission that clamping a membrane in a spraying process is obvious, but that if one of ordinary skill has the teaching that the membrane can be clamped, the specific type of clamp used for such purpose would be recognized by those skilled in the art. Appellant respectfully submits that the Examiner has not established a *prima facie* case of obviousness by saying it would be obvious to clamp the membrane without some teaching in the references of clamping or hold the membrane during the spraying process. Appellant's independent claim 1 includes spraying the catalyst on the membrane and clamping the membrane during the spraying process. In order to do this properly, the clamping must be done in a way that allows the catalyst ink to be properly deposited, such as by holding the membrane by its edge as shown in figure 1.

The Examiner states on page 4 of the Final Office Action that Starz does not disclose using a heat lamp for drying the membrane, but teaches a circulating air oven.

The Examiner dismisses the heat lamp limitation of Appellant's claims because no criticality has been given to the use of the heat lamp for drying the membrane.

Appellant respectfully submits that independent claim 13 includes the combination of spraying the catalyst ink on the membrane over several passes to deposit the ink on the membrane to the desired thickness, and drying the membrane under a heat lamp as the catalyst ink is being sprayed. Appellant submits that an air circulating oven would not be applicable for drying the membrane as multiple sprayed layers of the catalyst are being deposited on the membrane because the spraying apparatus would need to be inside of the oven. As the catalyst is being sprayed on the membrane, and is being dried at the same time, the catalyst can be dry before the next pass is sprayed on the previously deposited catalyst. Such a process increases the thickness control of the catalyst and the quality of the catalyst layer. Starz does not teach or suggest spraying the catalyst in multiple passes at the same time that the membrane is being dried. Therefore, Appellant respectfully submits that the Examiner has not established a *prima facie* case of obviousness concerning independent claim 13.

In the spraying step of independent claim 21, the ionomer in the catalyst that is sprayed on the membrane is about half of the ionomer concentration of the catalyst that is provided in the final cathode or anode of the MEA. Therefore, the other half of the ionomer comes from somewhere else, and not from the catalyst ink spray, such as by spraying an ionomer layer on the membrane prior to spraying the catalyst ink on the membrane as claimed in dependent claim 22.

The Examiner states on page 3 of the Final Office Action that the catalyst ink disclosed in Starz comprises a catalyst, a solvent and half the concentration of ionomer

in the final catalyst, referencing paragraphs [0031], [0032] and [0054]. Those paragraphs are provided below.

[0031] In the case of coating an ionomer membrane, the preparation with the nanoparticles, optionally mixed with other additives such as, for example, dissolved ionomer, carbon black or further electrocatalysts, is applied to the membrane in a spray process, by brushing or immersing or by means of screen printing. After coating, the temporary stabilizer is decomposed by treating with acid or alkali and it is then washed out. Dissolved ionomer is obtainable in aqueous solution with low molecular weight aliphatic alcohols (Fluka, Buchs; Aldrich, Steinheim). Aqueous solutions of the ionomer in higher concentrations (10 wt. % and 20 wt. %) can be prepared therefrom.

[0032] Ionomer membranes and also the ionomer contained in the catalyst layers can be used in an acidic proton-conducting H form or, after exchanging the protons for monovalent ions such as, for example, Na^+ and K^+ , in a non-acidic Na^+ or K^+ form for preparing membrane electrode assemblies. The non-acidic form of polymer membranes is usually more stable towards thermal stress than the acidic form and is therefore preferably used. Before using the membrane electrode assembly, however, the polymer electrolyte has first to be returned to its acidic, proton-conducting form. This is achieved by so-called reprotonation. Reprotonation is performed by treating the membrane electrode assembly in sulfuric acid.

[0054] 5.6 g of the colloidal solution (Pt content 1 wt. %) were dispersed with 0.4 g of an aqueous solution of Nafion (10 wt. % in water) and 0.1 g of carbon black (type: Vulcan XC-72, from Cabot) and the resulting ink was applied in a spray process to the front and rear faces of a Nafion membrane (type: Nafion 112, thickness 50 μm , from DuPont). Then drying was performed at temperatures of 80.degree. C. in a circulating air oven. The total Pt loading on the front and rear faces of the membrane was 0.2 mg Pt/cm². After drying, the catalyzed membrane was treated for 30 min in a sulfuric acid bath (0.5 normal, pH=0.3) and then washed with water. After that, it was placed between two non-catalyzed gas diffusion layers and incorporated into a PEM single cell.

Appellant has reviewed each of these paragraphs identified by the Examiner for teaching this limitation of Appellant's independent claim 21, as well as dependent claim 8, and can find no teaching or suggestion that the sprayed catalyst in Starz includes half

the concentration of ionomer in the final catalyst. Therefore, Appellant respectfully submits that the Examiner has not established a *prima facie* case of obviousness for independent claim 21.

MPEP 2143 provides a list of exemplary rationales that may support a conclusion of obviousness specifically:

(A) Combining prior art elements according to known methods to yield predictable results;

(B) Simple substitution of one known element for another to obtain predictable results;

(C) Use of known technique to improve similar devices (methods, or products) in the same way;

(D) Applying a known technique to a known device (method, or product) ready for improvement to yield predictable results;

(E) "Obvious to try" - choosing from a finite number of identified, predictable solutions, with a reasonable expectation of success;

(F) Known work in one field of endeavor may prompt variations of it for use in either the same field or a different one based on design incentives or other market forces if the variations are predictable to one of ordinary skill in the art;

(G) Some teaching, suggestion, or motivation in the prior art that would have led one of ordinary skill to modify the prior art reference or to combine prior art reference teachings to arrive at the claimed invention.

Rationale (A) states "prior art elements", rationale (B) states "one known element", rationale (C) states "known technique", rationale (D) refers to a "known technique to a known device", rationale (E) includes a "number of identified, predictable solutions", rationale (F) states "known work", and rationale (G) is the well developed,

teaching, suggestion and motivation requirement. Appellant respectfully submits that all these rationales contemplate the modification or analysis of a known element, technique, device, etc. Appellant respectfully submits that the several conclusions of obviousness by the Examiner for using a non-existing element in the prior art does not establish a proper case of *prima facie* obviousness. Appellant further submits that there is no teaching, suggestion or motivation in Starz for the clamping, multiple spraying steps, drying while spraying or the concentration of ionomer in the catalyst spray as discussed above.

4. Dependent claims 2, 5-8, 10, 11, 14 and 22-29

Several of the dependent claims provide various combinations of the elements discussed above to their respective in dependent claims, and therefore, in at least some circumstances provide methods that include a few of the claim elements wholly missing from Starz that the Examiner has held to be obvious. Appellant respectfully submits that the dependent claims also cannot be held obvious under the rationale suggested by the Examiner.

B. Claims 2, 6, 12-14, 22-24 and 29 are not obvious in view of Starz and Swathirajan

Swathirajan discloses processes for making electrodes for electro-chemical cells. The example identified in column 8 of Swathirajan provides a process for making a membrane electrode assembly 12. The Examiner has referred Appellant to column 9, lines 5-15 for teaching a heat lamp used to dry a catalyst layer. This section of Swathirajan talks about applying a layer of an AB slurry on a carbon sheet using a brush, doctor blade or spray gun, and then drying the sheet under a heat lamp. The AB slurry does include some platinum, but is not the main catalyst layer, rather is a barrier

layer. Column 9, lines 38-40 talks about applying a catalyst slurry by brushing and then drying at 100°C. No mention is made of spraying the catalyst slurry on the barrier layer and no mention is made of using a heat lamp to dry the catalyst slurry.

Regardless, Swathirajan does not disclose the step of independent claim 13, and other claims, of spraying the catalyst ink on the MEA over several passes at the same time the MEA is being dried under a heat lamp. Therefore, Appellant respectfully submits that the Examiner has not established a *prima facie* case of obviousness for claims 2, 6, 12-14, 22-24 and 29.

C. Claims 5, 12, 13, 23 and 29 are not obvious in view of Starz and Grot

Grot discloses a membrane and electrical structure, including a specific composition of particles in a catalyst ink. The Examiner has directed Appellant's attention to column 8, lines 15-50 of Grot as teaching using a heat lamp and pressure to fix a catalyst ink onto a membrane. This section of Grot states that the ink is fixed on the membrane by any one or a combination of "pressure, heat, adhesive, binder, solvent, electro-static and the like." No mention is made of spraying the ink on the membrane. Using heat and pressure to form a catalyst to a membrane is the type of operation that Appellant is attempting to avoid with their claimed invention because of the problems with using pressure, as discussed in Appellant's background discussion in paragraphs [0007] - [0014]. Column 8, line 47 of Grot mentions using a heat lamp to adhere the catalyst to the membrane. However, heating the membrane using heat lamps as disclosed by Grot does not include using heat lamps in combination with a catalyst spraying process including multiple spray passes. Therefore, Appellant submits that the Examiner has failed to provide a *prima facie* case of obviousness for the rejection of claims 5, 12, 13, 23 and 29 in view of Starz and Grot.

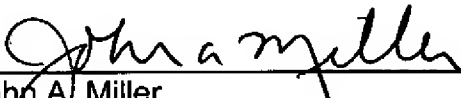
VIII. Conclusion

Appellant respectfully submits that claims 1, 2, 5-8, 10, 11, 13, 14 and 21-29 are not obvious in view of Starz, claims 2, 6, 12-14, 22-24 and 29 are not obvious in view of Swathirajan, and claims 5, 12, 13, 23 and 29 are not obvious in view of Starz and Grot. Therefore, it is respectfully requested that these rejections be withdrawn.

Respectfully submitted,

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CLAIMS APPENDIX

COPY OF CLAIMS INVOLVED IN THE APPEAL

This list of claims will replace all prior versions and listings of claims in the application.

1. A method for fabricating a membrane electrode assembly (MEA), said method comprising:

providing a proton conducting membrane in its protonated form having a first side and a second side;

spraying a catalyst ink on the first side of the membrane to deposit a catalyst layer of a cathode or an anode of the MEA; and

clamping the membrane in a clamp to prevent the membrane from wrinkling as a result of the wetness of the catalyst ink on the membrane.

2. The method according to claim 1 further comprising spraying an ionomer layer on the membrane prior to spraying the catalyst ink on the membrane.

5. The method according to claim 1 further comprising drying the MEA under a heat lamp to dry the catalyst layer.

6. The method according to claim 1 wherein spraying the catalyst ink includes spraying the ink over several passes to deposit the ink on the membrane to the desired thickness.

7. The method according to claim 1 further comprising spraying the catalyst ink on the second side of the membrane to deposit a catalyst layer of the other of the anode or the cathode.

8. The method according to claim 1 wherein the catalyst ink includes a catalyst, solvent and an ionomer having a concentration that is about half of the ionomer concentration of the catalyst as a ratio of ionomer to carbon in a final cathode or anode of the MEA.

10. The method according to claim 1 further comprising soaking the MEA in water.

11. The method according to claim 1 further comprising soaking the MEA in sulfuric acid to remove excess solvent and ensure protonation.

12. The method according to claim 1 further comprising hot-pressing the MEA after the catalyst ink is sprayed on the membrane to remove excess solvent and compress the catalyst layer.

13. A method for fabricating a membrane electrode assembly (MEA), said method comprising:

providing a proton conducting membrane in its protonated form;

spraying a catalyst ink on the membrane to deposit a catalyst layer of a cathode or an anode of the MEA, wherein spraying the catalyst ink includes spraying the

ink over several passes to deposit the ink on the membrane to the desired thickness;
and

drying the MEA under a heat lamp as the catalyst ink is being sprayed
during the several passes to dry the catalyst layer.

14. The method according to claim 13 further comprising spraying an ionomer layer on the membrane prior to spraying the catalyst ink on the membrane.

21. A method for fabricating a membrane electrode assembly (MEA), said method comprising:

providing a proton conducting membrane in its protonated form having a first side and a second side; and

spraying a catalyst ink on the first side of the membrane to deposit a catalyst layer of a cathode or an anode of the MEA, wherein the catalyst ink includes a catalyst, solvent and an ionomer having a concentration that is about half of the ionomer concentration of the catalyst as a ratio of ionomer to carbon in a final cathode or anode of the MEA.

22. The method according to claim 21 further comprising spraying an ionomer layer on the membrane prior to spraying the catalyst ink on the membrane.

23. The method according to claim 21 further comprising drying the MEA under a heat lamp to dry the catalyst layer.

24. The method according to claim 21 wherein spraying the catalyst ink includes spraying the catalyst ink over several passes to deposit the catalyst ink on the membrane to the desired thickness.

25. The method according to claim 21 further comprising spraying the catalyst ink on the second side of the membrane to deposit a catalyst layer of the other of the anode or the cathode.

26. The method according to claim 21 further comprising clamping the membrane in a clamp to prevent membrane wrinkling as a result of the wetness of the catalyst ink as it is being sprayed on the membrane.

27. The method according to claim 21 further comprising soaking the MEA in water.

28. The method according to claim 21 further comprising soaking the MEA in sulfuric acid to remove excess solvent and ensure protonation.

29. The method according to claim 21 further comprising hot-pressing the MEA after the catalyst ink is sprayed on the membrane to remove excess solvent and compress the catalyst layer.

EVIDENCE APPENDIX

There is no evidence pursuant to §1.130, §1.131 or §1.132.

RELATED PROCEEDINGS APPENDIX

There are no decisions rendered by a court or the Board in any proceeding identified in Section II of this Appeal Brief.